美國能源創新動態報導

DOE National Laboratories/
DOE Energy Innovation Hubs

No. 1　　2018 年 01 月 29 日

Solar-to-Fuel System Recycles CO2 to Make Ethanol and Ethylene

太陽能轉換燃料機制，回收二氧化碳並產出乙醇和乙烯

Berkeley Lab advance is first demonstration of efficient, light-powered production of fuel via artificial photosynthesis

Scientists at the Department of Energy’s Lawrence Berkeley National Laboratory (Berkeley Lab) have harnessed the power of photosynthesis to convert carbon dioxide into fuels and alcohols at efficiencies far greater than plants. The achievement marks a significant milestone in the effort to move toward sustainable sources of fuel.

本期摘要(KEY INFORMATION)

◎勞倫斯伯克利國家實驗室(Berkeley Lab)成功展示一種將二氧化碳直接轉換為乙醇和乙烯的方法，其能源轉換效率可媲美天然產品。研究人員藉由優化光伏電化學系統的每個組件來減少電壓損失，並在現有光伏系統不足時創建新材料。由於二氧化碳是頑固穩定的分子，將二氧化碳還原成如乙醇、乙烯等碳氫化合物最終產物可能需要高達 5 伏的電壓，但研究人員開發出可讓電壓減半的機制，同時保持產品的選擇性，並藉由在氧化鋅表面上增長氧化銥奈米管，打造特製的陽極，建立更均勻的表面積以支援較佳的化學反應。這項研究不僅對設計高效二氧化碳減排和新材料測試設備大有進展，且為全面整合太陽能驅動的二氧化碳減排設備的未來發展提供了清晰的架構。

◎芝加哥伊利諾斯大學(UIC)、人工光合作用聯合中心(JCAP)及勞倫斯伯克利國家實驗室(Berkeley Lab)的研究人員發現電觸媒藉由水和電將二氧化碳轉化為一氧化碳的方法，此有助於研發高效的電觸媒技術，俾利大規模生產合成氣，即一氧化碳和氫氣的混合物。以電化學方法將二氧化碳轉換為燃料，可說是提供了一種以化學鍵形式儲存風能、太陽輻射等能源的方法。研究團隊開發出一種最先進的多尺度模型，用以整合反應途徑的量子化學分析，以確切了解如何透過催化劑（在此使用的是銀）以電化學方法還原二氧化碳並將其製成一氧化碳。一旦理解電觸媒上的反應是如何發生的，即可控制催化劑的結構和操作條件，進而有效地產生一氧化碳。由於一氧化碳和氫氣不會溶解於含水電解質中，因此它們可輕易地以合成氣分離，並轉化成燃料，例如甲醇、二甲醚或烴類混合物。金屬中的銀藏量豐富且便宜，以大規模生產一氧化碳而言，是大有可為的電催化劑。

Many systems have successfully reduced carbon dioxide to chemical and fuel precursors, such as carbon monoxide or a mix of carbon monoxide and hydrogen known as syngas. This new work, described in a study published in the journal Energy and Environmental Science, is the first to successfully demonstrate the approach of going from carbon dioxide directly to target products, namely ethanol and ethylene, at energy conversion efficiencies rivaling natural counterparts.

The researchers did this by optimizing each component of a photovoltaic-electrochemical system to reduce voltage loss, and creating new materials when existing ones did not suffice.

"This is an exciting development," said study principal investigator Joel Ager, a Berkeley Lab scientist with joint appointments in the Materials Sciences and the Chemical Sciences divisions. "As rising atmospheric CO2 levels change Earth's climate, the need to develop sustainable sources of power has become increasingly urgent. Our work here shows that we have a plausible path to making fuels directly from sunlight."

That sun-to-fuel path is among the key goals of the Joint Center for Artificial Photosynthesis (JCAP), a DOE Energy Innovation Hub established in 2010 to advance solar fuel research. The study was conducted at JCAP's Berkeley Lab campus.

The initial focus of JCAP research was tackling the efficient splitting of water in the photosynthesis process. Having largely achieved that task using several types of devices, JCAP scientists doing solar-driven carbon dioxide reduction began setting their sights on achieving efficiencies similar to those demonstrated for water splitting, considered by many to be the next big challenge in artificial photosynthesis.

Another research group at Berkeley Lab is tackling this challenge by focusing on a specific component in a photovoltaic-electrochemical system. In a study published today, they describe a new catalyst that can achieve carbon dioxide to multicarbon conversion using record-low inputs of energy.

Not just for noon

For this JCAP study, researchers engineered a complete system to work at different times of day, not just at a light energy level of 1-sun illumination, which is equivalent to the peak of brightness at high noon on a sunny day. They varied the brightness of the light source to show that the system remained efficient even in low light conditions.

When the researchers coupled the electrodes to silicon photovoltaic cells, they achieved solar conversion efficiencies of 3 to 4 percent for 0.35 to 1-sun illumination. Changing the configuration to a high-performance, tandem solar cell connected in tandem yielded a conversion efficiency to hydrocarbons and oxygenates exceeding 5 percent at 1-sun illumination.

"We did a little dance in the lab when we reached 5 percent," said Ager, who also holds an appointment as an adjunct professor at UC Berkeley's Materials Science and Engineering Department.

Among the new components developed by the researchers are a copper-silver nanocoral cathode, which reduces the carbon dioxide to hydrocarbons and oxygenates, and an iridium oxide nanotube anode, which oxidizes the water and creates oxygen.

"The nice feature of the nanocoral is that, like plants, it can make the target products over a wide range of conditions, and it is very stable," said Ager.

The researchers characterized the materials at the National Center for Electron Microscopy at the Molecular Foundry, a DOE Office of Science User Facility at Berkeley Lab. The results helped them understand how the metals functioned in the bimetallic cathode. Specifically, they learned that silver aids in the reduction of carbon dioxide to carbon monoxide, while the copper picks up from there to reduce carbon monoxide further to hydrocarbons and alcohols.

Seeking better, low-energy breakups

Because carbon dioxide is a stubbornly stable molecule, breaking it up typically involves a significant input of energy.

“Reducing CO2 to a hydrocarbon end product like ethanol or ethylene can take up to 5 volts, start to finish,” said study lead author Gurudayal, postdoctoral fellow at Berkeley Lab. “Our system reduced that by half while maintaining the selectivity of products.”

Notably, the electrodes operated well in water, a neutral pH environment.

“Research groups working on anodes mostly do so using alkaline conditions since anodes typically require a high pH environment, which is not ideal for the solubility of CO2,” said Gurudayal. “It is very difficult to find an anode that works in neutral conditions.”

The researchers customized the anode by growing the iridium oxide nanotubes on a zinc oxide surface to create a more uniform surface area to better support chemical reactions.

“By working through each step so carefully, these researchers demonstrated a level of performance and efficiency that people did not think was possible at this point,” said Berkeley Lab chemist Frances Houle, JCAP deputy director for Science and Research Integration, who was not part of the study. “This is a big step forward in the design of devices for efficient CO2 reduction and testing of new materials, and it provides a clear framework for the future advancement of fully integrated solar-driven CO2-reduction devices.”

Other co-authors on the study include James Bullock, a Berkeley Lab postdoctoral researcher in materials sciences, who was instrumental in engineering the system’s photovoltaic and electrolysis cell pairing. Bullock works in the lab of study co-author Ali Javey, Berkeley Lab senior faculty scientist and a UC Berkeley professor of electrical engineering and computer sciences.

This work is supported by the DOE Office of Science.

原始連結：

http://newscenter.lbl.gov/2017/09/18/solar-fuel-system-recycles-co2-for-ethanol-ethylene/

Converting Carbon Dioxide to Carbon Monoxide Using Water, Electricity

用水、電將二氧化碳轉化為一氧化碳

Researchers at the University of Illinois at Chicago and the Joint Center for Artificial Photosynthesis have determined how electrocatalysts can convert carbon dioxide to carbon monoxide using water and electricity. The discovery can lead to the development of efficient electrocatalysts for large scale production of synthesis gas — a mixture of carbon monoxide and hydrogen.

“The electrochemical reduction of carbon dioxide to fuels is a subject of considerable interest because it offers a means for storing electricity from energy sources such as wind and solar radiation in the form of chemical bonds,” said Meenesh Singh, assistant professor of chemical engineering and lead author on the study published in the journal Proceedings of the National Academy of Sciences.

During his postdoctoral research at the University of California, Berkeley, Singh studied artificial photosynthesis and was part of a team that developed artificial leaves that, when exposed to direct sunlight, were capable of converting carbon dioxide to fuels.

In his latest research, Singh developed a state-of-the-art multiscale model that unites a quantum-chemical analysis of reaction pathway; a microkinetic model of the reaction dynamics; and a continuum model for transport of species in the electrolyte to learn precisely how carbon dioxide can be electrochemically reduced through a catalyst, in this case silver, and made into carbon monoxide.

While the most plausible reaction pathway is usually identified from quantum-chemical calculation of the lowest free-energy pathway, this approach can be misleading when coverages of adsorbed species differ significantly, Singh said. It is essential, therefore, to integrate the effects from electronic states of a catalyst at the atomic-level with the dynamics of species in the electrolyte at the continuum-level for accurate prediction of electrocatalytic reaction pathways.

“This multiscale model is one of the biggest accomplishments in electrochemistry,” he said.

To understand how electrocatalysts in fuel cells or electrochemical cells work, scientists need to first probe the electronic and quantum levels, which can be extremely challenging in the presence of an electric field, said Jason Goodpaster, assistant professor of chemistry at the University of Minnesota and one of the co-authors. It took Singh and Goodpaster more than one year to individually produce and benchmark the models and integrate them into a multiscale framework for full-scale simulation of the electrochemical reaction.

This is the first time, Singh said, that scientists have predicted quantitatively from first principles, the current density of carbon monoxide and hydrogen as a function of applied potential and pressure of carbon dioxide.

“Once you recognize how these reactions are occurring on electrocatalysts, you can control the catalysts structure and operating conditions to produce carbon monoxide efficiently,” Singh said. Since they are product gases — carbon monoxide and hydrogen are insoluble in aqueous electrolytes — they can be readily separated as synthesis gas and converted into fuels such as methanol, dimethyl ether, or a mixture of hydrocarbons.

Electrocatalysts such as gold, silver, zinc, palladium and gallium are known to yield mixtures of carbon dioxide and hydrogen at various ratios depending on the applied voltage, Singh said. Gold and silver exhibit the highest activity towards carbon dioxide reduction, and since silver is more abundant and less expensive than gold, “silver is the more promising electrocatalyst for large-scale production of carbon monoxide,” he said.

Along with Goodpaster, co-authors include Adam Weber, Lawrence Berkeley National Laboratory, Berkeley, Calif.; and Martin Head-Gordon and Alexis Bell, University of California, Berkeley. This work was performed at UIC’s Materials and Systems Engineering Laboratory and the Joint Center for Artificial Photosynthesis, a Department of Energy (DOE) Energy Innovation Hub, supported through the Office of Science of the U.S. Department of Energy under Award DE-SC0004993. UIC also provided financial support.

原始連結：https://today.uic.edu/converting-carbon-dioxide-to-carbon-monoxide-using-water-electricity